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IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF :
HELMUT BREHM, ET AL. : EXAMINER: H. L. PEZZUTO
SERIAL NO: 10/069,721 :
FILED: FEBRUARY 28, 2002 : GROUP ART UNIT: 1713
FOR: POLYMERIZATE COMPOSITION :
AND A METHOD FOR PRODUCING THE
SAME

REPLY BRIEF

ASSISTANT COMMISSIONER FOR PATENTS
WASHINGTON, D.C. 20231

SIR:

Responsive to the Examiner's Answer of January 27, 2006, please consider the following:

The Examiner has adhered to the rejection of Claims 1-7 and 19-20 under 35 U.S.C. § 102(e) as being anticipated by or, in the alternative, under 35 U.S.C. §103 (a) as being obvious over Patel et al (US 6,103,839). In addition, the Examiner has maintained the rejection of Claims 1-7, 9-17 and 19-20 under 35 U.S.C. §§ 102(b), (e) as being anticipated by or, in the alternative, under 35 U.S.C. §103 (a) as being obvious over Hatsuda et al, EP 0 630 909 A1 or EP 0 296 331 B1. These rejections are again traversed.

Notably, the Examiner asserts that no evidence has been provided of the actual difference in properties or that the claimed process steps inherently and necessarily result in a different product. See page 8 of Examiner's Answer. Applicants disagree.

Applicants wish to point out that the specification provides substantial evidence that the

structure and properties of polymers depend on how the polymerization is performed and that different polymerization processes result in different polymers, having different structures and properties.

The water-soluble polymer composition according to claim 1 is obtained by a continuous polymerization wherein during said polymerization at least one parameter biasing the polymerization is varied according to a recurrent pattern. Preferably, at least one of the following parameters biasing the polymerization is varied:

- a) the composition of the monomer solution,
- b) the concentration of the monomer solution,
- c) the amount of catalyst,
- d) the amount of molecular weight modifier, and/or
- e) the pH value of the monomer solution

(present application, page 4, 4th paragraph).

It is a feature of claim 1 of the present application, however, that **said at least one parameter is not varied arbitrarily**. In contrast, **it is required that the parameter is varied according to a recurrent pattern**. This means that the parameter biasing the polymerization is varied in a desired manner, but at regularly recurring time intervals within a reasonable range familiar to those skilled in the art, and preferably in a continuous fashion. The pattern preferably is an oscillation about a mean value selectable at random. Said oscillation preferably is harmonic or anharmonic and preferably undamped. Amplitude and frequency of the oscillation can be selected at random. While the frequency, i.e., the time during which the pattern of varying a polymerization parameter is carried out once, is determined by the sizing of the plant components, the amplitude, i.e., the level of the continuously performed variation, is crucial for the application-technical properties of the polymer composition (present application, page 4, 2nd and 3rd paragraph; page 5, 2nd paragraph).

The following considerations further explain that the variation of a parameter biasing the polymerization according to a recurrent pattern indeed directly and inevitably effects the properties of the product obtained from said polymerization.

a) According to the fundamental concepts of polymer chemistry the kinetic chain length $\bar{\nu}$ is correlated to the degree of polymerization of the polymer formed, at a given time. Under stationary state conditions in chain-growth polymerization the kinetic chain length $\bar{\nu}$ is proportional to the monomer concentration $[M]$ divided by the square root of the initiator concentration $[I]$, i.e.

$$\bar{\nu} \propto \frac{[M]}{\sqrt{I}}.$$

Therefore, decreasing the initiator concentration $[I]$ results in an increase of $\bar{\nu}$ and vice versa. In consequence, an oscillation of the initiator concentration causes an oscillation of $\bar{\nu}$ which in turn **corresponds to an oscillation of the degree of polymerization (molecular weight)**.

The above is illustrated in the following: assuming the following given set of conditions at a given time

- an initiator concentration $[I]$ of 1.0 mol^{-1} results in an average degree of polymerization of about 1,000 and
- an initiator concentration $[I]$ of 0.1 mol^{-1} results in an average degree of polymerization of about 5,000.

Thus, when oscillating the initiator concentration $[I]$ within the limits 1.0 mol^{-1} and 0.1 mol^{-1} , the degree of polymerization of the polymers will oscillate within the limits of 1,000 and 5,000.

It is evident that the polymer composition obtained by such a polymerization process, i.e. the final product obtained over a long period of time and a certain number of

oscillations, exhibits a relatively broad molecular weight distribution when compared with the polymer compositions obtained from a process wherein the initiator concentration is kept constant.

b) Another illustration is the copolymerization of the monomers A and B. For example, if monomer A is capable of reacting with another monomer A or with monomer B and if there is no preference in terms of reactivity. Thus, the probability of whether the growing polymer chain reacts with either monomer A or monomer B only depends upon the individual concentrations of the two monomers, i.e. [A] and [B].

When the concentrations of monomer A and monomer B are identical ($[A]=[B]$), the polymerization yields a product statistically incorporating an equal amount of monomer units A and B. Such product may be exemplified by a mixture of the following three individualized polymers (12 monomer units incorporated in a polymer chain, 6 units of monomer A and 6 units of monomer B):

AABABBABABBA

ABBAAABABBAB

BAABABBABBA

When the concentration of monomer A is lower than the concentration of monomer B ($[A]<[B]$), the polymerization would yield a product statistically incorporating less monomer units A than monomer units B. Such product being enriched in monomer units B may be exemplified by a mixture of the following three individualized polymers (12 monomer units incorporated in a polymer chain, 3 units of monomer A and 9 units of monomer B):

BABABBBBABBB

ABBBBABABBBB

BBABBBBBBBAA.

On the other hand, when the concentration of monomer A is higher than the concentration of monomer B ($[A]>[B]$), the polymerization would yield a product statistically incorporating more monomer units A than monomer units B. Such product being enriched in monomer units A may be exemplified by a mixture of the following three individualized

polymers (12 monomer units incorporated in a polymer chain, 9 units of monomer A and 3 units of monomer B):

AAAABAABAABA AABAAABAABAA BAABAABAAAAA.

Summing up, the polymer compositions obtained under these conditions are displayed in the table here below:

concentr.	representative polymers			ratio A:B
[A]=[B]	AABABBABABBA	ABBAAABABBAB	BAABABBABBAA	1:1
[A]<[B]	BABABBBBABBB	ABBBBABABBBB	BBABBBBBBBAA	1:3
[A]>[B]	AAAABAABAABA	AABAAABAABAA	BAABAABAAAAA	3:1

Therefore, when oscillating the concentration of monomer A and keeping the concentration of monomer B constant, a polymer composition is obtained comprising

- polymers with a balanced content of monomer units A and B,
- polymers enriched in monomer units B, and
- polymers enriched in monomer units A.

In contrast, however, when not performing an oscillation of the monomer concentration, a polymer composition is obtained only comprising polymers of a constant statistical distribution of monomers A and B.

This mode of varying at least one parameter biasing the polymerization reaction according to a recurrent pattern is further illustrated in the specification of the present application.

In a preferred embodiment of the process according to the invention, a constant amount of a catalyst solution is metered into the constant feed flow of an acrylamide solution purged

with nitrogen. A solution of a cationic monomer is metered from another reservoir in an amount continuously varying in the form of a sine-shaped oscillation (cf. present application, page 9, 2nd paragraph).

When comparing this embodiment with the above copolymerization of monomers A and B, acrylamide corresponds to monomer B (constant concentration) and the cationic monomer corresponds to monomer A (oscillating concentration).

As a result of the exothermic reaction, the varying monomer concentration gives rise to a continuously changing temperature in the polymer product on the polymerization belt. Samples taken at the end of the dryer at intervals of 5 minutes clearly show the varying cationic properties oscillating about a mean value (cf. present application, page 9, 2nd paragraph), as a polymer being enriched in monomer units A (cationic monomer) exhibits a higher degree of ionogenicity than a polymer being enriched in monomer units B (acrylamide monomer).

c) It is also demonstrated in **example 2 of the present application** that there is a direct correlation between the parameters of the process and the properties of the polymers obtained therefrom.

In example 2 acrylamide (ACA) is copolymerized with dimethylaminopropyl acrylamide quaternized with methyl chloride (DIMAPA quat., cationic monomer). The metering rate of DIMAPA quat. is varied with a steady increase from 12.8 kg/h to 95 kg/h and with a steady decrease back to 12.8 kg/h over a time period of 60 minutes. This recurrent pattern of varying amounts is maintained throughout the entire test period.

Following a residence time of 45 minutes, the solid polymer gel is ground and dried. Samples are taken at the end of the dryer at intervals of 5 minutes. These individual fractions of the overall product exhibit viscosities ranging from about 425 mPa·s to about 500 mPa·s and ionogenicities ranging from about 45% to about 59% (cf. present application, page 17, table).

The overall product, i.e. the mixture of all fractions, has an average viscosity of 470 mPa·s and an average ionogenicity of 53%.

It is emphasized that the time period of a single oscillation of the metering rate of DIMAPA quat. (60 minutes) is longer than the residence time of the polymers in the continuous polymerization process (45 minutes) and that the oscillation of the metering rate of DIMAPA quat. indeed causes the concentration of this monomer within the reactor to vary according to a recurrent pattern (oscillation about a mean value).

It becomes evident from the comparative data summarized in the table on page 21 of the present application that the polymer composition obtained from the process according to example 2 is superior over the polymer compositions of the prior art which have been obtained from conventional polymerization processes.

The most relevant data are displayed in the table here below:

composition	monomers	overall viscosity	overall ionogenicity	concentration	consumption	thickened sludge	setting degree
example 2	ACA + DIMAPA	460 mPa·s	54%	5 g/l	1.31	33%	98.6%
Praestol®	ACA + DIMAPA	470 mPa·s	53%	5 g/l	1.32	26.9%	98.7%

The polymer composition of the invention is found to achieve improved sludge thickening (33% instead of 26.9%) with unchanged settling degree and polymer addition.

d) Similar experimental evidence that indeed there is a direct correlation between the parameters of the process and the properties of the polymers obtained there from is provided in **example 3 of the present application.**

In example 3 acrylamide (ACA) is also copolymerized with dimethylaminopropyl acrylamide quaternized with methyl chloride (DIMAPA quat., cationic monomer). The metering rate of ACA is varied with a steady increase from 17 kg/h to 120 kg/h and with a

steady decrease back to 17 kg/h over a time period of 60 minutes. This recurrent pattern of varying amounts is maintained throughout the entire test period.

The polymer gel obtained after 40 minutes (residence time) is ground and dried. Samples are taken at the end of the dryer at intervals of 5 minutes. These individual fractions of the overall product exhibit viscosities ranging from about 325 mPa·s to about 460 mPa·s and ionogenicities ranging from about 49% to about 57.7% (cf. present application, page 17, table). The overall product, i.e. the mixture of all fractions, has an average viscosity of 350 mPa·s and an average ionogenicity of 53.0%.

It becomes evident from the comparative data summarized in the table on page 20 of the present application that the polymer composition obtained from the process according to example 3 is superior over the polymer compositions of the prior art which have been obtained from conventional polymerization processes.

The most relevant data are displayed in the table here below.

composition	monomers	overall viscosity	overall ionogenicity		added amount [g/m ³]							
					160	170	180	190	200	220	260	280
example 3	ACA + DIMAPA	350 mPa·s	53%	time	29	25	18					
				clarity	26	27	29					
Praestol® 644 BC	ACA + DIMAPA	365 mPa·s	52%	time				27	24	16		
				clarity				31	33	34		
Praestol® 650 BC	ACA + DIMAPA	370 mPa·s	39%	time							50	31
				clarity							26	30

The polymer composition of the invention is found to permit removal of solids with virtually constant filtrate clarity and comparable rate, but less polymer employed (160-180 g/m³).

Patel et al (US 6,103,839) fail to disclose or suggest a water-soluble **polymer composition** obtained by continuous polymerization of at least one unsaturated monomer, wherein during said polymerization **at least one parameter** biasing the polymerization is

varied according to a recurrent pattern. As a result, the polymer composition according to the present invention is different from the polymers of Patel et al (US 6,103,839).

The control over molecular weight distribution and temperature and continuous feeding of monomers, initiators, chain transfer agents etc. disclosed in Patel et al is **not a variation according to a recurrent pattern** of at least one parameter biasing the polymerization. In other words, just because a polymerization is continuous or certain parameters are controlled it does not mean that a parameter is varied according to a recurrent pattern. Applicants filed schematic figures illustrating examples of recurrent patterns according to the present invention with the Amendment of October 14, 2004. In addition, Applicants filed a scheme showing in a simplified manner a continuous addition of monomer as described in Patel et al. The differences are clear from the schematic drawings.

With regard to the product-by-process claims (Claims 1-7 and 20), Applicants wish to point out that the **structure and properties of polymers depend on how the polymerization is performed**. Different polymerization processes result in different polymers, having different structures and properties. Since the processes of Patel et al and the present invention are different, the claimed polymer composition is different from the polymers of Patel et al.

Regarding Claim 19, Patel et al (US 6,103,839) fail to disclose or suggest a water-soluble **polymer composition** obtained by continuous polymerization of at least one unsaturated monomer, wherein during said polymerization **at least one parameter** biasing the polymerization is **varied according to a recurrent pattern**. In particular, fail to disclose or suggest that a parameter **oscillates** about a mean value during the polymerization. In other words there are no recurring minima and maxima of a parameter such as those claimed in Claim 19:

- a concentration of at least one monomer,
- an amount of a catalyst,

- an amount of a molecular weight modifier,
- a pH value of a monomer solution, or
- a composition of said monomer solution.

As a result, the polymer of Claim 19 is different from the polymers of the cited reference.

Further, the polymer composition according to the present invention is different from the polymers of Hatsuda et al, or EP 0 630 909 A1 or EP 0 296 331 B1 because they fail to disclose or suggest a water-soluble **polymer composition** obtained by continuous polymerization of at least one unsaturated monomer, wherein during said polymerization **at least one parameter** biasing the polymerization is varied according to a recurrent pattern. Applicants wish to point out that the **structure and properties of polymers depend on how the polymerization is performed**. Different polymerization processes result in different polymers, having different structures and properties. Since the processes of Hatsuda et al, EP 0 630 909 A1 or EP 0 296 331 B1 and the present invention are different (Claims 9-17), the claimed polymer composition (Claims 1-7, 15, 16, 19 and 20) is different from the polymers of these prior art references.

The Examiner argues that Hatsuda et al embraces the recurrent pattern as claimed because the temperature will vary between 60-95°C (Office Action of May 5, 2005, page 4, last paragraph). According to the Examiner, this meets the claimed requirement of a parameter **biasing** according to regular reoccurring time integrals. However, Applicants disagree.

The controlling of the maximum attained temperature of the polymerization system in the range of from 60-95°C disclosed in Hatsuda et al (Abstract) is **not a variation according to a recurrent pattern** of at least one parameter biasing the polymerization. In Hatsuda et al, the polymerization heat is merely removed by using both cooling due to heat conduction transfer and cooling due to the latent heat of vaporization (col. 2, lines 44-48). Hatsuda et al do

nothing to vary the temperature according to a recurrent pattern.

Further, the Examiner argues that EP 0630 909 shows drop wise addition of monomer (Office Action of May 5, 2005, page 5, 1st paragraph). According to the Examiner, this meets the claimed requirement of a parameter **biasing** according to regular reoccurring time integrals. However, Applicants disagree. EP 0 630 909 A1 merely discloses feeding monomer in increments, but there is no **recurrent pattern** in which parameters biasing the polymerization are **varied at regular recurring time intervals**.

EP 0 630 909 A1 discloses at page 5, lines 41 to 42 that the reaction mixture is added during the polymerization incrementally or otherwise. Applicants disagree with the Examiner's allegation that "incrementally" means necessarily "dropwise." In addition, "incrementally" does not mean that the monomer is added according to a recurrent pattern as claimed. There is simply no disclosure in EP 0 630 909 A1 that drops are added according to a recurrent pattern as claimed. Applicants filed a scheme illustrating in a simplified manner incremental addition of monomer as disclosed in this reference with the Amendment of October 14, 2004. Since the processes of EP 0 630 909 A1 and the present invention are different (Claim 9-17), the claimed polymer composition (Claims 1-7, 15, 16 19 and 20) is different from the polymers of EP 0 630 909 A1.

With regard to EP 0 296 331 B1 the Examiner refers to the specification, the paragraph bridging pages 8 and 9 (Office Action of May 5, 2005, page 5, 2nd paragraph). However, contrary to the Examiner's position, **this paragraph in the specification describes that *the continuous polymerization of EP 0 296 331 B1 is modified to carry out a process according to the present invention:***

"To carry out said process variant, the continuous polymerization described in EP 0 296 331, Example 4 and Fig. 2 **is modified** in such a way that the mass flow of catalyst solution is varied by a regulator via metering valves 28 and 30 according to a preselected pattern at regular time intervals in a recurring fashion."

Specification, paragraph bridging pages 8 and 9. Emphasis added.

Thus, the process of EP 0 296 331 B1 is different from the claimed process and the resulting polymers are different from the claimed polymers.

Regarding Claim 19, Hatsuda et al, or EP 0 630 909 A1 or EP 0 296 331 B1 fail to disclose or suggest a water-soluble **polymer composition** obtained by continuous polymerization of at least one unsaturated monomer, wherein during said polymerization **at least one parameter** biasing the polymerization is varied according to a recurrent pattern. In particular, these prior art references fail to disclose or suggest that a parameter **oscillates** about a mean value during the polymerization. In other words there are no recurring minima and maxima of a parameter such as those claimed in Claim 19:

- a concentration of at least one monomer,
- an amount of a catalyst,
- an amount of a molecular weight modifier,
- a pH value of a monomer solution, or
- a composition of said monomer solution.

As a result, the polymer of Claim 19 is different from the polymers of the cited references.

Finally, the Examiner has adhered to the provisional double patenting rejection of Claims 1-7, 9-17 and 19-20 over claims 13-15, 17-19 and 26 of copending application Serial No. 10/069,278, now US 6,911,499.

The claims of US 6,911,499 are directed polymer compositions that **absorb aqueous fluids**, while the present claims are directed to polymers that are water-soluble. Notably, the polymer compositions of US 6,911,499 are powdered and cross-linked which makes them useful as polymers which adsorb aqueous fluids. See Claims 1 and 7 of US 6,911,499. Thus,

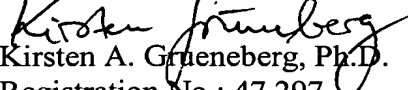
the polymers of US 6,911,499 are not water-soluble as required by the claims of the present invention.

For all the above reasons, it is respectfully requested that the rejections of the appealed claims be REVERSED.

Respectfully submitted,

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